

## DECLARATION

I, Naeun Jun, a Korean citizen of 101-201, Kyungnam Anusvill, Sinsu-dong, Mapo-gu, Seoul, Korea do hereby solemnly and sincerely declare as follows:

1. That I am well acquainted with the English and Korean languages.
2. That the following is a correct translation into English of Korean Patent Application No. 2003-45323 filed July 4, 2003, and I make the solemn declaration conscientiously believing the same to be true.

Seoul, June 20, 2006

*N. E. Jun*

---

Naeun Jun

[DOCUMENT] Application for patent  
[CATEGORY OF RIGHT] Patent  
[RECEIVING PERSON] The commissioner of the Korean Intellectual Property Office  
[DATE OF FILING] 2003. 07. 04

[TITLE OF THE INVENTION-KOREAN] 나프탈렌테트라카르복시디이미드 유도체  
및 이를 포함하는 전자사진감광체

[TITLE OF THE INVENTION-ENGLISH] Naphthalenetetracarboxylic acid diimide  
derivatives and electrophotographic photoconductive material

[APPLICANT]

[NAME] Samsung Electronics Co., Ltd.

[APPLICANT CODE] 1-1998-104271-3

[EMPOWERED ATTORNEY]

[NAME-KOREAN] 정홍식

[ATTORNEY CODE] 9-1998-000543-3

[REGISTRATION NUMBER OF GENERAL POWER OF ATTORNEY]

2003-002208-1

[INVENTOR]

[NAME-KOREAN] 김승주

[NAME-ENGLISH] KIM,SEUNG JU

[RESIDENT REGISTRATION NUMBER] 720810-1675611

[ZIP CODE] 442-726

[ADDRESS] 912-1106, Byeokjeokgol 9-danji Apartment,  
Youngtong-dong, Paldal-gu, Suwon-city, Gyunggi-do

[NATIONALITY] KR

[INVENTOR]

[NAME-KOREAN] 요코다 사부로

[NAME-ENGLISH] YOKOTA,SABURO

[ADDRESS] 554-1202 Jinheung Apartment, Youngtong-dong,  
Paldal-gu, Suwon-city, Gyunggi-do

[NATIONALITY] JP

[INVENTOR]

[NAME-KOREAN] 연경열

[NAME-ENGLISH] YON,KYUNG YOL

[RESIDENT REGISTRATION NUMBER] 630324-1042129

[ZIP CODE] 463-050

[ADDRESS] 508-1104, Samhwan Apartment, Hyoja-chon, 301,  
Seohyeon-dong, Bundang-gu, Seongnam-city,  
Gyeonggi-do

[NATIONALITY] KR

[INVENTOR]

[NAME-KOREAN] 이환구

[NAME-ENGLISH] LEE,HWAN KOO

[RESIDENT REGISTRATION NUMBER] 670923-1056925

[ZIP CODE] 440-040

[ADDRESS] 147-2, Sinpung-dong, Jangan-gu, Suwon-city,  
Gyeonggi-do

[NATIONALITY] KR

[INVENTOR]

[NAME-KOREAN] 김범준

[NAME-ENGLISH] KIM,BEOM JUN

[RESIDENT REGISTRATION NUMBER] 700502-1019313

[ZIP CODE] 463-773

[ADDRESS] 212-202, Wooseong Apartment, Sibeom-danji,  
Seohyeon-dong, Bundang-gu, Seongnam-city,  
Gyeonggi-do

[NATIONALITY] KR

[REQUEST FOR EXAMINATION] Requested

[PURPOSE] I, hereby, submit the present application for the Patent under the Article 42 and  
request Examination under the Article 60 of the Patent Law.

Agent Hong-sik JEONG (seal affixed)

[Official Fee]

[Basic fee] 20 pages 29,000 won

[Additional fee] 11 pages 11,000 won

[Claiming Priority Right]	0	case	0 won
[Filing Request For Examination]	11	claims	461,000 won
[Total]			501,000 won

[Enclosed Documents]    An abstraction and specification (drawing)

5

10

15

20

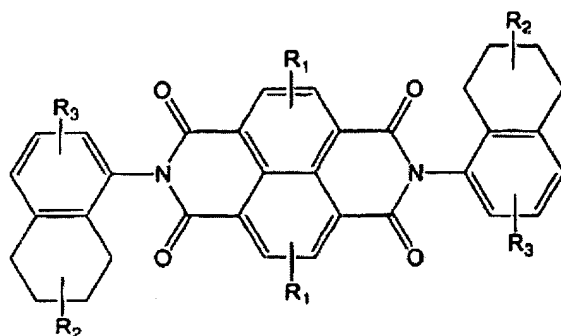
25

**[ABSTRACT]**

**[Abstract of the disclosure]**

Disclosed are naphthalenetetracarboxylic acid diimide derivatives and an electrophotographic photoconductive material comprising the same. The naphthalenetetracarboxylic acid diimide derivatives have a structure represented by the following formula:

in which, R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> are each independently any one selected from the group consisting of hydrogen; halogen; substituted or unsubstituted alkyl of C<sub>1</sub> to C<sub>20</sub>; substituted or unsubstituted alkoxy of C<sub>1</sub> to C<sub>20</sub>; substituted or unsubstituted aryl of C<sub>6</sub> to C<sub>30</sub>; and substituted or unsubstituted aralkyl of C<sub>7</sub> to C<sub>30</sub>. The electrophotographic photoconductive material comprises the naphthalenetetracarboxylic acid diimide derivatives as an electron transferring material. Accordingly, it is possible to provide an electrophotographic photoconductive material having electron transferring ability improved by using the novel naphthalenetetracarboxylic acid diimide derivatives as an electron transferring material.



**[Search terms]**

Naphthalenetetracarboxylic acid diimide, photoconductive material

## **[SPECIFICATION]**

### **[The title of the invention]**

Naphthalenetetracarboxylic acid diimide derivatives and electrophotographic photoconductive material

### **5 [Detailed description of the invention]**

#### **[Object of the invention]**

#### **[The field of the invention and the prior art]**

The present invention relates to naphthalenetetracarboxylic acid diimide derivatives and an electrophotographic photoconductive material comprising the same. More particularly, it relates to naphthalenetetracarboxylic acid diimide derivatives comprising a specific substituent and an electrophotographic photoconductive material having improved electron transporting ability by using the same as an electron transferring material.

In an electrophotographic image forming apparatus, a photoconductive material is electrically charged and exposed to an image forming light source to form an electrostatic latent image. Next, the latent image is developed by a toner by applying a development voltage. The developed toner image is transferred to a recording medium such as paper, followed by fixation of the transferred image. Such electrophotography is widely used in various apparatuses such as digital or analogue copying machines, printers, facsimiles, etc.

As a photoconductive material of the electrophotographic image forming apparatus, selenium photoconductive materials, amorphous silicone photoconductive materials, etc. were used in the past, but now, organic photoconductive materials are widely used. The organic photoconductive materials are classified into a multi-layered photoconductive material, in which a charge generating material (CGM) and a charge transferring material (CTM) are separately distributed into different layers of a layered laminate and perform different functions, and a single-layer photoconductive material, in which a charge generating

material and a charge transferring material are dispersed in a single layer. The multi-layered photoconductive material is mainly used to produce a (-)type organic photoconductive material, while the single-layer photoconductive material is mainly used to produce (+)type organic photoconductive material.

5           The (+)type single-layer organic photoconductive material has advantages that it generates less ozone harmful to human bodies and it can be produced at a relatively low production cost since it has a single layer structure.

          In the (+)type organic photoconductive material, a photosensitive layer comprises an electron transferring material, a hole transferring material, a binder resin and a charge  
10   generating material. Since the electron transferring ability of the common electron transferring materials presently used is 100 times less than the hole transferring ability of the hole transferring material, the performance of the organic photoconductive material is largely affected by the electron transferring ability of the electron transferring material. Therefore, among the components contained in the photosensitive layer of the (+)type organic  
15   photoconductive material, the electron transferring material is the most important.

          Examples of compounds commonly used as the electron transferring material include dicyanofluorenone, 2-nitro-9- fluorenone, 2,7-dinitro-9- fluorenone, 2,4,7-trinitro-9-  
fluorenone, 2,4,5,7-tetranitro-9- fluorenone, 2-nitrobenzothiopene, 2,4,8-trinitrothioxanthrone, dinitroanthracene, dinitroacridine, dinitroanthraquinone, naphthoquinone and 3,5-dimethyl-  
20   3'5'-di-t-butylidiphenquinone. Since compounds having the dicyanofluorenone and diphenquinone structures has a weak electron transferring ability, when an organic photoconductive material is produced using these compounds as an electron transferring material, there are problems such as reduction in the charged potential and increase in the exposure potential upon use of a long period of time.

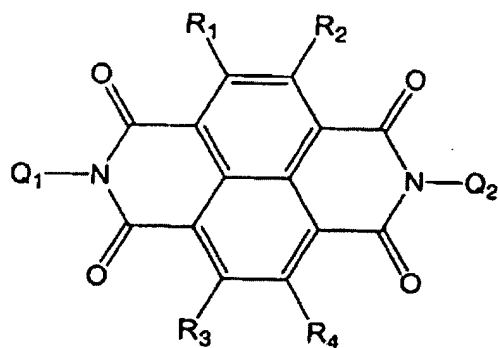
25           The electron transferring material having the naphthalenetetracarboxylic acid diimide



structure is known to have an electron transferring ability superior to the naphthoquinone structure. The electron transferring ability of the electron transferring material having the naphthalenetetracarboxylic acid diimide structure is largely affected by solubility of the electron transferring material in an organic solvent and compatibility of the electron transferring material with a polymer binder resin.

US Pat. NO. 4,442,193 disclosed a photoconductive material composition comprising a photoconductive substance and a 1,4,5,8-naphthalenebisdicarboxylic acid diimide derivative compound.

US Pat. NO. 5,468,583 disclosed an photoconductive material comprising a conductive layer, a charge generating layer and a polymer binder having an electron transferring material dispersed therein, in which the electron transferring material comprises at least one of cyclic bisdicarboxylic acid imide compounds represented by the following formula I:



in which, Q<sub>1</sub> and Q<sub>2</sub> are independently branched chain alkyl, un-substituted straight chain alkyl, unsubstituted cycloalkyl, alkyl-substituted cycloalkyl, unsubstituted straight-chain unsaturated alkyl, aryl, C<sub>2</sub> to C<sub>20</sub> alkyl, alkoxy or hydrogen, provided that both Q<sub>1</sub> and Q<sub>2</sub> are not hydrogen, R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub> and R<sub>4</sub> are, independently, hydrogen, C<sub>2</sub> to C<sub>4</sub> alkyl, a C<sub>2</sub> to C<sub>4</sub> alkoxy or halogen.

All of the electron transferring materials disclosed in the above patent comprise

naphthalenetetracarboxylic acid diimide derivatives as an electron transferring material.

The naphthalenetetracarboxylic acid diimide derivatives are the most expected compounds as an electron transferring material owing to their electron transferring abilities. However, the naphthalenetetracarboxylic acid diimide derivatives known so far are not sufficiently  
5 satisfactory in solubility to an organic solvent and compatibility with an used binder resin. Also, since they are apt to crystallize and form precipitates in an photosensitive layer and an electron transferring layer, electrophotographic properties of the photoconductive material may be deteriorated.

Meanwhile, when naphthalenetetracarboxylic acid diimide derivative compounds are  
10 used in a concentration which can maintain compatibility with a binder resin, the photoconductivity of the photoconductive material may be deteriorated.

#### **[Technical object of the invention]**

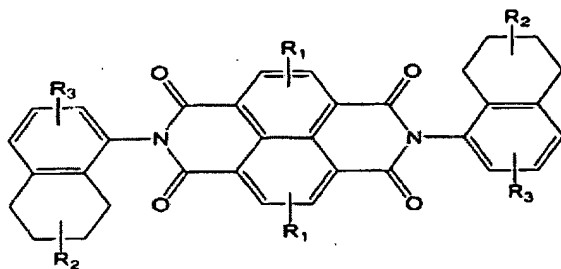
The present invention has been made to solve the above problems and thus, one aspect of the present invention is to provide novel naphthalenetetracarboxylic acid diimide  
15 derivative compounds with improved electron transferring ability.

It is another aspect of the present invention to provide an electrophotographic photoconductive material using a novel naphthalenetetracarboxylic acid diimide derivative compound having high solubility in an organic solvent, excellent compatibility with a binder resin and improved electron transferring ability as an electron transferring material.

#### **[Construction of the invention]**

  
20

In order to achieve the above objects, the naphthalenetetracarboxylic acid diimide derivatives according to the present invention can be represented by the following formula (II),

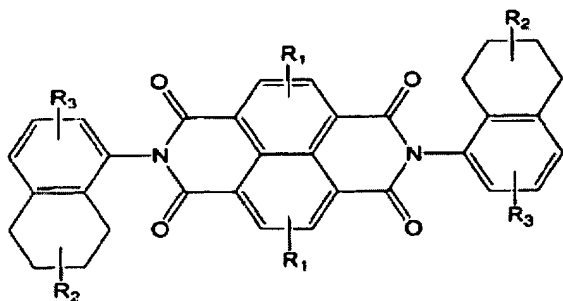


in which,  $R_1$ ,  $R_2$ , and  $R_3$  are, independently, any one selected from the group consisting of hydrogen; halogen; substituted or unsubstituted alkyl of  $C_1$  to  $C_{20}$ ; substituted or unsubstituted alkoxy of  $C_1$  to  $C_{20}$ ; substituted or unsubstituted aryl of  $C_6$  to  $C_{30}$ ; and substituted or unsubstituted aralkyl of  $C_7$  to  $C_{30}$ .

The substituted alkyl, substituted alkoxy and substituted aralkyl may be independently substituted with any one selected from the group consisting of alkyl, aryl, halogen and alkoxy.

The substituted aryl may be substituted with any one selected from the group consisting of alkyl, alkoxy, nitro and halogen.

Also, in order to achieve the above objects, the electrophotographic photoconductive material according to the present invention comprises a substrate and a photosensitive layer formed on the substrate, in which the photosensitive layer comprises a charge generating material and a charge transferring material, and the charge transferring material comprises, as an electron transferring material, a naphthalenetetracarboxylic acid diimide derivative represented by the following formula (III),



wherein, R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> are each independently any one selected from the group consisting of hydrogen; halogen atom; substituted or unsubstituted alkyl of C<sub>1</sub> to C<sub>20</sub>; substituted or unsubstituted alkoxy of C<sub>1</sub> to C<sub>20</sub>; substituted or unsubstituted aryl of C<sub>6</sub> to C<sub>30</sub>; and substituted or unsubstituted aralkyl of C<sub>7</sub> to C<sub>30</sub>.

5           The substituted alkyl, substituted alkoxy and substituted aralkyl may be independently substituted with any one selected from the group consisting of alkyl, aryl, halogen and alkoxy.

          The substituted aryl may be substituted with any one selected from the group consisting of alkyl, alkoxy, nitro and halogen.

10           Preferably, the electrophotographic photoconductive material contains the electron transferring material of the above formula (III) in a ratio of 20wt% to 40wt% based on the total solid components.

          The electrophotographic photoconductive material may further comprise a hole transferring material.

15           The photosensitive layer of the electrophotographic photoconductive material has a single-layer structure comprising the charge generating material and the charge transferring material which are dispersed in a binder resin.

          Also, the photosensitive layer of the electrophotographic photoconductive material may have a multi-layered structure comprising a charge generating layer comprising the  
20   charge generating material and a charge transferring layer comprising the charge transferring material.

          The electrophotographic photoconductive material may further comprise an electron acceptor.

          Now, the present invention will be described in detail.

25           The naphthalenetetracarboxylic acid diimide derivatives according to the present

invention represented by the formula (II) have a tetrahydronaphthalene structure attached to nitrogen atoms at both ends of a naphthalene-1,4,5,8-tetracarboxylic acid diimide structure. Referring to the Examples described below, the naphthalenetetracarboxylic acid diimide derivatives containing the tetrahydronaphthalene group may be synthesized.

5           The naphthalenetetracarboxylic acid diimide derivatives have a structural represented by the formula (II), in which  $R_1$ ,  $R_2$ , and  $R_3$  are each independently any one selected from the group consisting of hydrogen; halogen; substituted or unsubstituted alkyl of  $C_1$  to  $C_{20}$ ; substituted or unsubstituted alkoxy of  $C_1$  to  $C_{20}$ ; substituted or unsubstituted aryl of  $C_6$  to  $C_{30}$ ; and substituted or unsubstituted aralkyl of  $C_7$  to  $C_{30}$ .

10           In the formula (II), where  $R_1$ ,  $R_2$ , and  $R_3$  are alkyl groups, they may be  $C_1$  to  $C_{20}$  straight or branched alkyl groups. Particularly,  $C_1$  to  $C_{12}$  straight or branched alkyl groups are preferred. Usable alkyl group includes, for example, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, t-butyl, pentyl, hexyl, 1,2-dimethyl-propyl, 2-ethyl-hexyl and the like, but are not limited thereto.

15           In the formula (II), where  $R_1$ ,  $R_2$ , and  $R_3$  are alkoxy groups, they may be  $C_1$  to  $C_{20}$  alkoxy groups. Usable alkoxy group includes methoxy, ethoxy, propoxy, phtoxy, pentyloxy and the like, but are not limited thereto.

          In the formula, where  $R_1$ ,  $R_2$ , and  $R_3$  are aryl groups, they may be a  $C_6$  to  $C_{30}$  aryl groups. Examples of usable aryl group include phenyl, tolyl, biphenyl, o-terphenyl, 20 naphthyl, anthryl, phenanthryl and the like, but are not limited thereto.

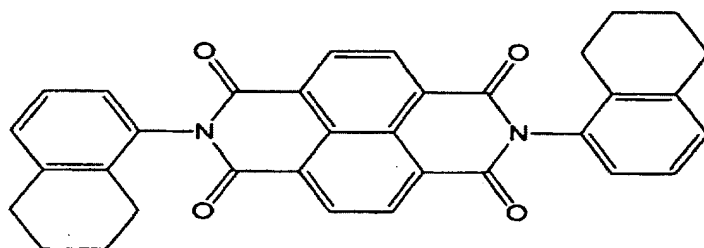
          In the formula, where  $R_1$ ,  $R_2$ , and  $R_3$  are aralkyl groups, they may be a  $C_7$  to  $C_{30}$  aralkyl groups. The term "aralkyl group" used herein refers collectively complex groups having a formula of  $Ar(CH_2)_n-$ , which are formed by substitution of aromatic hydrocarbon group (aryl group), such as phenyl, tolyl and the like, at a carbon atom of an alkyl group, and 25 is an abbreviation for arylalkyl group. Usable aralkyl group includes, for example, benzyl

(C<sub>6</sub>H<sub>5</sub>CH<sub>2</sub>-), phenethyl(C<sub>6</sub>H<sub>5</sub>CH<sub>2</sub>CH<sub>2</sub>-), and the like, but are not limited thereto.

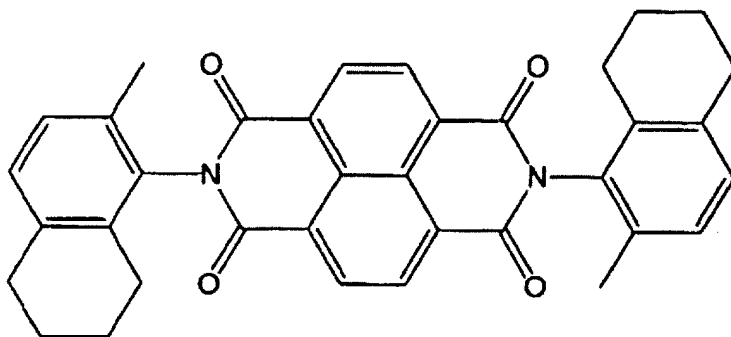
The alkyl group, alkoxy group and aralkyl group may independently have a substituent of alkyl such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, t-butyl, pentyl, hexyl, 1,2-dimethyl-propyl and 2-ethyl-hexyl; halogen such as fluorine, chlorine, bromine and iodine; aryl such as phenyl, tolyl, biphenyl, o-terphenyl, naphthyl, anthryl and phenanthryl; or alkoxy such as methoxy, ethoxy, propoxy, phtoxy and pentyloxy, but the usable substituent is not limited thereto.

The aryl group may have a substituent of alkyl such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, t-butyl, pentyl, hexyl, 1,2-dimethyl-propyl and 2-ethyl-hexyl; halogen such as fluorine, chlorine, bromine and iodine; nitro; or alkoxy such as methoxy, ethoxy, propoxy, phtoxy and pentyloxy; but the usable substituent is not limited thereto.

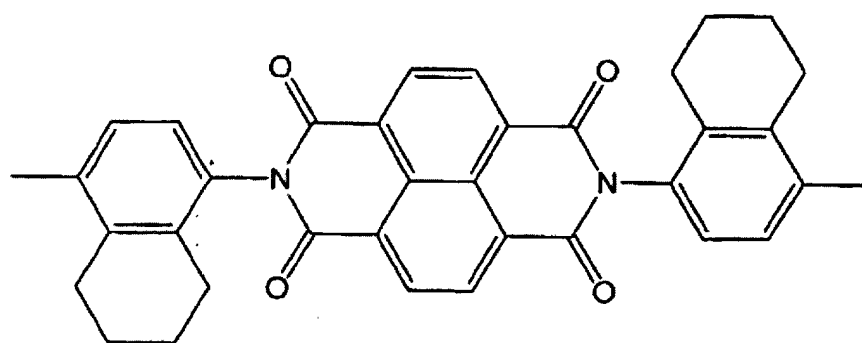
Examples of the naphthalenetetracarboxylic acid diimide derivatives according to the present invention as described above are as follows, but are not limited thereto.



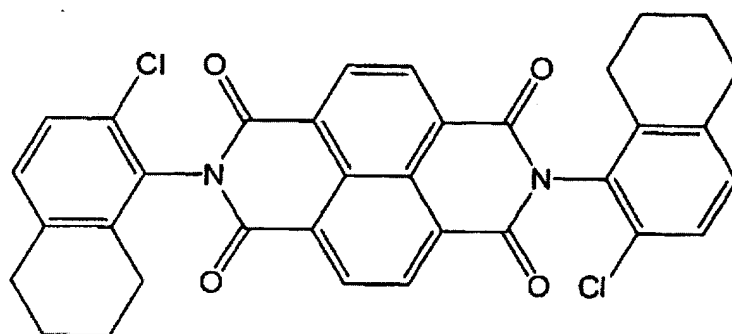
(IV)



(V)

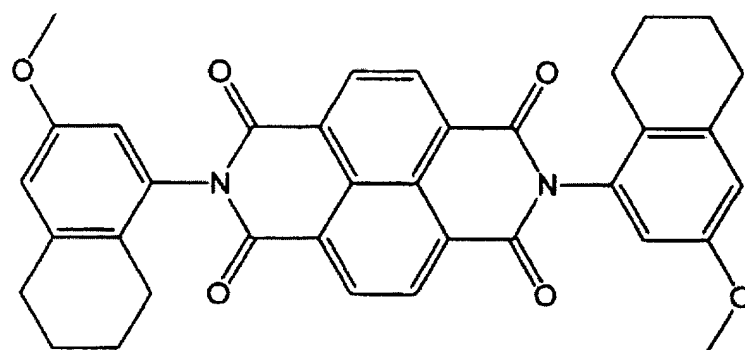


(VI)

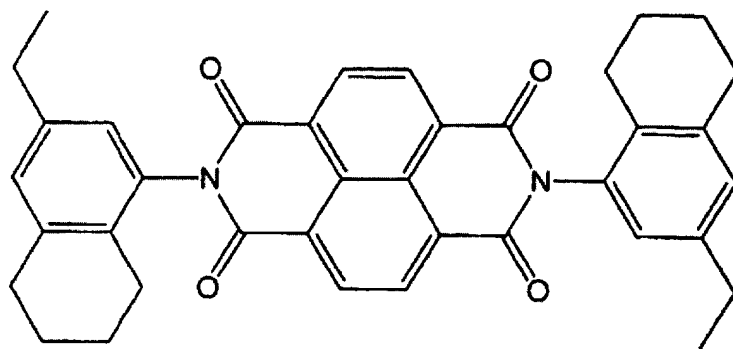


(VII)

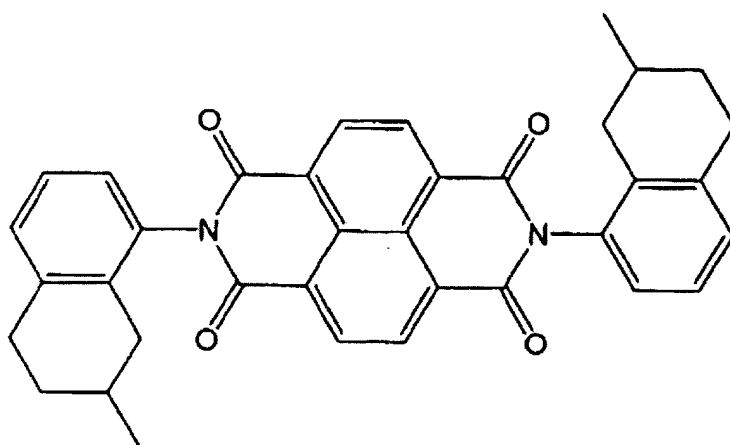
5



(VIII)



(IX)

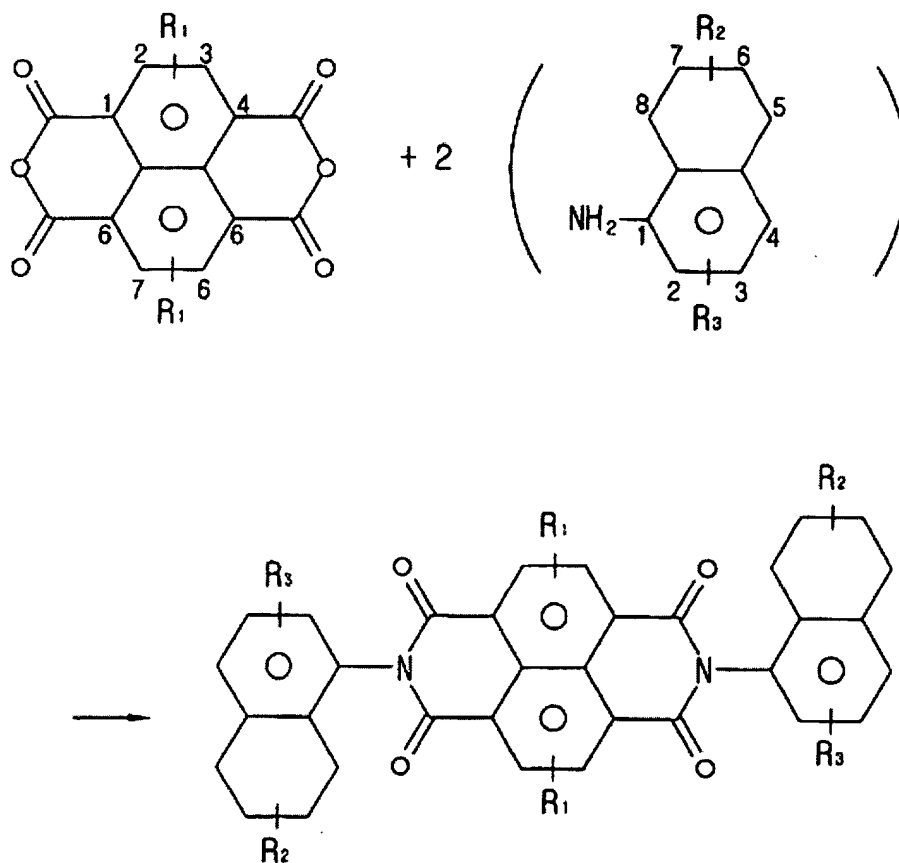


In order to synthesize the naphthalenetetracarboxylic acid diimide derivative of the  
 5 formula (II), amino-tetrahydronaphthalene(amino-tetrahydronaphthalene) is reacted with  
 naphthalenetetracarboxylic dianhydride.

An representative reaction scheme for synthesis of the naphthalenetetracarboxylic  
 acid diimide derivative according to the present invention is shown as follows.

Reaction Scheme (I)





As shown in the Reaction scheme 1, naphthalene-1,4,5,8-tetracarboxylic dianhydride may be reacted with 5,6,7,8-tetrahydro-1-naphthylamine to synthesize the naphthalenetetracarboxylic acid diimide derivatives.

The electrophotographic photoconductive material according to the present invention may be prepared by the following embodiment.

The electrophotographic photoconductive material comprises a substrate and a photosensitive layer formed on the substrate, in which the photosensitive layer comprises a charge generating material and a charge transferring material.

The substrate should be made of a conductive material. Examples of materials which can be used as the substrate include metals such as aluminum, copper, tin, platinum, gold, silver, vanadium, molybdenum, chrome, cadmium, titanium, nickel, indium, stainless

steel or brass; plastics having the foregoing metals deposited or laminated thereon; or glass coated with aluminum iodide, tin oxide or indium oxide and particularly, an aluminum substrate is preferred.

The charge generating material contained in the photosensitive layer may be largely  
5 classified into an inorganic charge generating material and an organic charge generating material. The inorganic charge generating material may include, for example, zinc oxide, lead oxide and selenium. The organic charge generating material may include various pigment materials including phthalocyanine type pigments, various soluble organic compounds including organometals and polymeric organic charge generating materials.

10 Examples of the organic charge generating material include azo pigments, disazo pigments, anthanthrone pigments, phthalocyanine pigments, indigo pigments, threne pigments, toluidine pigments, pyrazoline pigments, phenylene pigments, quinacridone pigments and the like, which may be used alone or in combination. Particularly, phthalocyanine pigments, perylene pigments and bisazo pigments are preferably used.

15 Examples of the phthalocyanine pigments include non-metal phthalocyanine, aluminum phthalocyanine, vanadium phthalocyanine, cadmium phthalocyanine, antimony phthalocyanine, chrome phthalocyanine, copper 4-phthalocyanine, germanium phthalocyanine, zinc phthalocyanine, chloroaluminum phthalocyanine, oxotitanyl phthalocyanine, chloroindium phthalocyanine, chlorogallium phthalocyanine, magnesium  
20 phthalocyanine, dialkyl phthalocyanine, tetramethyl phthalocyanine, and tetraphenyl phthalocyanine, but are not limited thereto.

The charge transferring material contained in photosensitive layer may be largely classified into a hole transferring material and an electron transferring material. The charge transferring material which is used in the present invention is an electron transferring material,  
25 which is the naphthalenetetracarboxylic acid diimide derivatives, as described above.

The charge transferring material may further comprise a hole transferring material along with the electron transferring material. Examples of the hole transferring material include poly-N-vinylcarbazole, phenanthrene, N-ethylcarbazole, 2,5-diphenyl-1,3,4-oxadiazole, 2,5-bis-(4-diethylaminophenyl)-1,3,4-oxadiazole, bis-didiethylaminophenyl-1,3,6-oxadiazole, 4,4'-bis(diethylamino)-2,2'-dimethyltriphenylmethane, 2,4,5-triaminophenylimidazole, 2,5-bis(4-diethylaminophenyl)-1,3,4-triazole, 1-phenyl-3-(4-diethylaminostyryle)-5-(4-diethylaminophenyl)-2-pyrazoline, tetra(m-methylphenyl)methaphenylenediamine, N,N,N',N'-tetraphenylbenzidine derivatives, N,N'-diphenyl-N,N'-disilylbenzidine and the like, but are not limited thereto.

The charge generating material and the charge transferring material contained in the photosensitive layer are dispersed in the binder resin. Examples of the binder resin include styrene-butadiene copolymers; polyvinyl toluene-styrene copolymers; silicone resins, styrene alkyd resins, silicone-alkyd resins; soya-alkyd resins; poly(vinyl chloride); poly(vinylidene chloride); vinylidene chloride-acrylonitrile copolymers; poly(vinylacetate); vinyl acetate-vinylchloride copolymers; poly(vinylacetal) such as poly(vinylbutyral); polyacrylic- and methacrylic ester such as poly(methylmethacrylate), poly(n-butylmethacrylate) and poly(isobutylmethacrylate); polystyrene, nitrated polystyrene; polymethylstyrene; isobutylene polymers; polyester such as poly[4,4'-(2-norborvinylidene)bisphenylene azelate-co-terephthalate (60/40)] and poly[ethylene-co-alkylene-bis(alkylene-oxyaryl)-phenylenedicarboxylate]; phenolformaldehyde resins; ketone resins; polyamides; polycarbonates; polythiocarbonates; poly[ethylene-co-isopropylidene-2,2-bis(ethyleneoxyphenylene)terephthalate]; copolymers of vinylhaloarylate and vinyl acetate such as poly(vinyl-m-bromobenzoate-co-vinyl acetate); polyolefine such as polyethylene chloride; and equivalents thereof. Particularly, polyester and polycarbonate are preferably used.

Preferably, the electrophotographic photoconductive material according to the present invention contains the electron transferring material of the formula (III) in a ratio of 20wt% to 40wt% based on the total weight of solid components. If the content of the electron transferring material is less than 20wt%, the electron transferring ability of the electron transferring material is deteriorated, while if the content exceeds 40wt% the solubility of the electron transferring material in a solvent is lowered or the compatibility with a binder resin is deteriorated.

The photosensitive layer comprising the electron transferring material of the formula (III) may be of a single layer structure or a laminate having a multi-layered structure.

The photosensitive layer comprising the electron transferring material of the formula (III) may be of a single layer structure or a laminate having a multi-layered structure.

The electrophotographic photoconductive material according to the present invention may further comprise an electron acceptor in combination with the electron transferring material. Usable electron acceptors include, for example, benzoquinone type compounds such as p-benzoquinone and 2,6-t-butylbenzoquinone; naphthoquinone type compounds such as 1,4-naphthoquinone, 2-t-butyl-3-benzoyl-1,4-naphthoquinone and 2-phenyl-3-benzoyl-1,4-naphthoquinone; and diphenoquinone type compounds such as 3,5-dimethyl-3'5'-di-t-butyl diphenoquinone, 3,5-dimethoxy-3',5',di-t-butyl diphenoquinone, 3,3'-dimethyl-5,5'-di-t-butyl diphenoquinone, 3,5'-dimethyl-3'5'-di-t-butyl diphenoquinone, 3,5,3',5'-tetramethyl diphenoquinone, 2,6,2',6'-tetra-t-butyl diphenoquinone, 3,5,3',5'-tetraphenyl diphenoquinone and 3,5,3',5'-tetraacetyldiphenoquinone, but are not limited thereto.

The electrophotographic photoconductive material comprises essentially a conductive substrate and a photosensitive layer formed on the substrate. The photosensitive layer is classified into a single-layer type photosensitive layer and a multi-layered type photosensitive

layer according to the layer number in the photosensitive layer and hence, the electrophotographic photoconductive material is also classified into a single-layer type electrophotographic photoconductive material and a multi-layered type electrophotographic photoconductive material. The single-layer type photosensitive layer and the multi-layered type photosensitive layer are differed by whether the charge generating layer and the charge transferring layer play their roles without distinction in a single layer or they are separated into different layers according to their roles, thereby forming a laminate. However, it should be understood that the lamination of a photosensitive layer on a conductive substrate to form a photoconductive material is identically applied to all the embodiments according to the present invention.

The lamination of the photosensitive layer on the substrate is performed by preparing a coating solution comprising the charge generating material and the charge transferring material, as described above, and coating the coating solution on the substrate. The coating solution is prepared using an organic solvent. Examples of organic solvents which can be used in the present invention include alcohols such as methanol, ethanol, isopropanol and butanol; aliphatic hydrocarbon compounds such as n-hexane, octane and cyclohexane; aromatic hydrocarbon compounds such as benzene, toluene, and xylene; halogenated hydrocarbon compounds such as dichloromethane, dichloroethane, carbon tetrachloride and chlorobenzene; ethers such as dimethyl ether, diethyl ether, tetrahydrofuran, ethyleneglycol dimethyl ether and diethyleneglycol dimethyl ether; ketone compounds such as acetone, methyl ethyl ketone and cyclohexane; ester compounds such as ethyl acetate and methyl acetate; and dimethylformamide, dimethyl sulfoxide and the like, in which the set forth compounds may be used alone or in combination of two or more thereof.

When an aromatic is directly attached to an imide structure of the naphthalenetetracarboxylic acid diimide derivatives, the plane of benzene and the plane of

naphthalene cross each other. Therefore, the two planes are oriented almost perpendicular to each other due to steric hinderance. That is, since the molecular structure of the naphthalenetetracarboxylic acid diimide derivative is not present in a single plane and hardly form an overlapped structure, the molecules are seldom crystallized with each other.

5 However, though the crystallization between molecules is difficult, the coupling of naphthalenetetracarboxylic acid diimide derivative with the organic solvent molecule may become easy. Therefore, the solubility of the naphthalenetetracarboxylic acid diimide derivative to the organic solvent may increase.

Also, the tetrahydronaphthalene structure of the naphthalenetetracarboxylic acid  
10 diimide derivatives according to the present invention, which comprises benzene attached to an aliphatic cyclic compound, does not spatially form a plane but a zigzag line. For an aliphatic cyclic compound, a chair conformation, that is, a trans-type isomer, is 3-dimensionally stable. The aliphatic ring attached to benzene of the tetrahydronaphthalene structure according to the present invention has a zigzag folded conformation and thus, is  
15 structurally stable. Since the zigzag folded structure is asymmetric and the aliphatic cyclic compounds are structurally flexible, the coupling with the solvent molecule may be increased by the asymmetry and flexibility. Such ziazag folded molecular structure can be expected using chem 3D Ultra 6.0 program.

Therefore, the naphthalenetetracarboxylic acid diimide derivatives having the  
20 tetrahydronaphthalene structure according to the present invention have increased solubility in organic solvents and improved compatibility with binder resins.

Examples according to the present invention and effects thereof will be provided below.

{Examples}

## Synthesis of naphthalenetetracarboxylic acid diimide derivatives

### Example 1

Reference was made to the Reaction Scheme (I).

10.72g (0.04mol) of naphthalene-1,4,5,8-tetracarboxylic acid dianhydride was mixed with 100ml of dimethylformamide (DMF) at room temperature while stirring. To the stirred solution, a mixture of 12.37g (0.084mol) of 5,6,7,8-tetrahydro-1-naphthylamine and 20ml of DMF was slowly added and stirred for 1 hour at room temperature. The reaction was refluxed for 3 hours while elevating the temperature, followed by cooling to room temperature.

To the cooled solution, 60ml of methanol was added to form precipitates, followed by filtration. The filtered solids were recrystallized from a solvent mixture of chloroform and ethanol.

The recrystallized product was dried in vacuo to obtain 20.0g of yellow crystals with a yield of 95%.

### **Preparation of electrophotographic photoconductive material**

#### Example 2

Electron transferring material of the formula (IV):	4.5 weight parts
---	------------------

$\alpha$ -type TiOPC of the following formula (XI):	0.9 weight parts
---	------------------

Hole transferring material of the following formula (XII)	9 weight parts
---	----------------

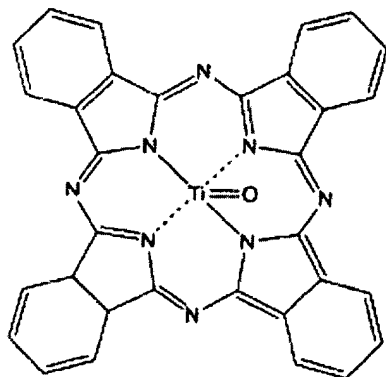
Binder resin of the following formula (XIII):	15.9 weight parts
---	-------------------

Methylene chloride:	84 weight parts
---------------------	-----------------

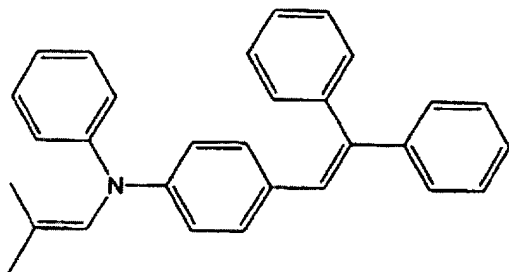
1,1,2-trichloroethane:	36 weight parts
------------------------	-----------------

The ingredients in the above weight ratio were sandmilled for 2 hours and dispersed by ultrasonic agitation. Then, the dispersion was coated on an aluminum-PET sheet by ring coating and dried at 110°C for 1 hour. Thus, an electrophotographic photoconductive

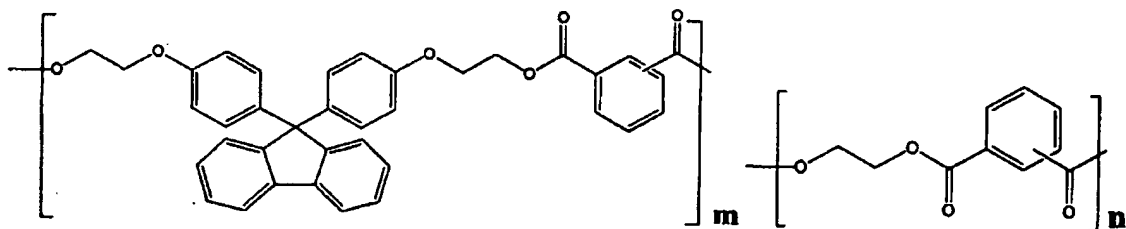
material having a thickness of about 12  $\mu\text{m}$  was prepared.



(XI)



(XII)



(XIII)

### **Example 3**

Electron transferring material of the formula (IV): 4.05 weight parts

$\alpha$  -type TiOPC of the formula (XI): 0.9 weight parts

Hole transferring material of the formula (XII): 9 weight parts

Binder resin of the formula (XIII): 15.9 weight parts

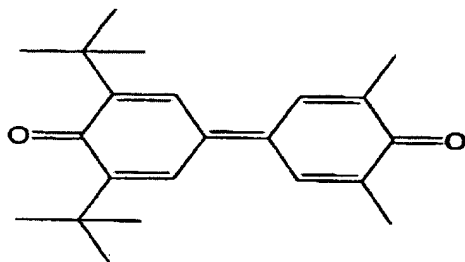


Methylene chloride: 84 weight parts

1,1,2-trichloroethane: 36 weight parts

Electron acceptor of the following formula (XIV): 0.45 weight parts

The ingredients in the above weight ratio were sandmilled for 2 hours and dispersed  
5 by ultrasonic agitation. Then, the dispersion was coated on an aluminum-PET sheet by ring  
coating and dried at 110°C for 1 hour. Thus, an electrophotographic photoconductive  
material having a thickness of about 12  $\mu\text{m}$  was prepared.



(XIV)

### Comparative Example 1

10 Electron transferring material of the following formula (XV): 4.5 weight parts

$\alpha$  -type TiOPC of the formula (XI): 0.9 weight  
parts

Hole transferring material of the formula (XII): 9 weight parts

Binder resin of the formula (XIII): 15.9 weight parts

15 Methylene chloride: 84 weight  
parts

1,1,2-trichloroethane: 36 weight  
parts

The ingredients in the above weight ratio were sandmilled for 2 hours and dispersed



parts

Hole transferring material of the formula (XII): 9 weight parts

Binder resin of the formula (XIII): 15.9 weight parts

Methylene chloride: 84 weight

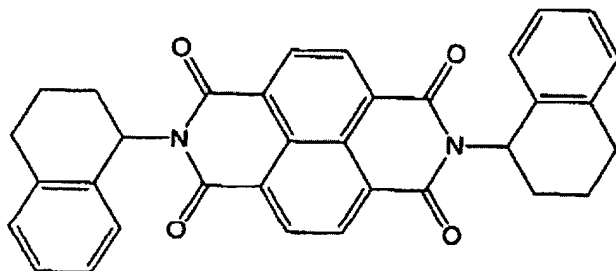
5 parts

1,1,2-trichloroethane: 36 weight

parts

The ingredients in the above weight ratio were sandmilled for 2 hours and dispersed by ultrasonic agitation. Then, the dispersion was coated on an aluminum-PET sheet by ring

10 coating and dried at 110°C for 1 hour.



(XVI)

#### **Comparative Example 4**

Electron transferring material of the formula (XVI): 4.05 weight parts

15

$\alpha$  -type TiOPC of the formula (XI): 0.9 weight parts

Hole transferring material of the formula (XII): 9 weight parts

Binder resin of the formula (XIII): 15.9 weight parts

Methylene chloride: 84 weight parts

1,1,2-trichloroethane: 36 weight parts

20

Electron acceptor of the formula (XIV): 0.45 weight parts

The ingredients in the above weight ratio were sandmilled for 2 hours and dispersed by ultrasonic agitation. Then, the dispersion was coated on an aluminum-PET sheet by ring coating and dried at 110°C for 1 hour.

5            **Comparative Example 5**

$\alpha$ -type TiOPC of the formula (XI):	0.9 weight parts
Hole transferring material of the formula (XII):	13.5 weight parts
Binder resin of the formula (XIII):	15.9 weight parts
Methylene chloride:	84 weight parts
10        1,1,2-trichloroethane:	36 weight parts

The ingredients in the above weight ratio were sandmilled for 2 hours and dispersed by ultrasonic agitation. Then, the dispersion was coated on an aluminum-PET sheet by ring coating and dried at 110°C for 1 hour.

15           **Comparative Example 6**

$\alpha$ -type TiOPC of the formula (XI):	0.9 weight parts
Hole transferring material of the formula (XII):	13.05 weight parts
Binder resin of the formula (XIII):	15.9 weight parts
Methylene chloride:	84 weight parts
20        1,1,2-trichloroethane:	36 weight parts
Electron acceptor of the formula (XIV):	0.45 weight parts

The ingredients in the above weight ratio were sandmilled for 2 hours and dispersed by ultrasonic agitation. Then, the dispersion was coated on an aluminum-PET sheet by ring

coating and dried at 110°C for 1 hour.

{Evaluation}

Potentials of the electrophotographic photoconductive material prepared in Example 2 to Comparative Example 6 are shown in Table 1 below.

5 Table 1

	V <sub>0</sub>	V <sub>d</sub>	V <sub>0</sub> 100	V <sub>d</sub> 100
Example 2	470	91	470	92
Example 3	487	91	489	94
Comp. Example 1	477	107	415	97
Comp. Example 2	481	105	438	100
Comp. Example 3	444	101	397	93
Comp. Example 4	490	96	452	92
Comp. Example 5	485	112	440	104
Comp. Example 6	494	113	448	102

In the table, V<sub>0</sub> represents initial charged potential, V<sub>d</sub> represents initial exposure potential, V<sub>0</sub> 100 represents charged potential after 100 cycles, and V<sub>d</sub> 100 represents exposure potential after 100 cycles.

10 As can be seen from Table 1, Example 2 and Example 3 showed lower initial exposure potentials as compared to Comparative Example 1 to Comparative Example 6. Therefore, it was noted that they are more efficient because electric power needed upon exposure is reduced.

Also, in Example 2 and Example 3, it was shown that the charged potential and  
15 exposure potential after 100 cycles were similar to those at the initial stage without any significant difference, that is, the charged potential and exposure potential were maintained

uniformly. However, in Comparative Examples 1 to 6, it was shown that the charged potential and exposure potential after 100 cycles were significantly lowered, as compared to those at the initial stage.

Therefore, it was noted that the electrophotographic photoconductive material comprising the electron transferring material according to the present invention has excellent electron transferring ability and the electron transferring ability can be maintained for a long period of time.

**[Effect of the invention]**

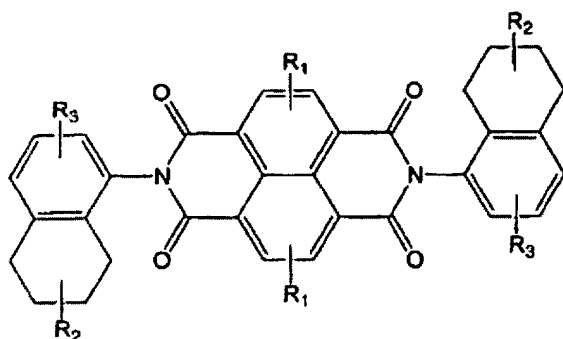
As described above, according to the present invention, it is possible to provide novel naphthalenetetracarboxylic acid diimide derivatives having increased solubility in organic solvents and improved compatibility with binder resins. Also, it is possible to produce an electrophotographic photoconductive material with excellent electron transferring ability by using the naphthalenetetracarboxylic acid diimide derivatives.

So far, although the present invention has been illustrated and explained in detail by preferred embodiments described above, it should be understood that the present invention is not limited thereto, various modifications and changes can be made by those skilled in the art, without departing from the scope and spirit of the invention as disclosed in the accompanying claims and such modifications and changes fall in the scope of the present invention.

# WHAT IS CLAIMED IS:

## [Claim 1]

A naphthalenetetracarboxylic acid diimide derivative represented by the following  
 10 formula:



in which, R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> are each independently any one selected from the group  
 consisting of hydrogen; halogen; substituted or unsubstituted alkyl of C<sub>1</sub> to C<sub>20</sub>; substituted or  
 unsubstituted alkoxy of C<sub>1</sub> to C<sub>20</sub>; substituted or unsubstituted aryl of C<sub>6</sub> to C<sub>30</sub>; and  
 15 substituted or unsubstituted aralkyl of C<sub>7</sub> to C<sub>30</sub>.

## [Claim 2]

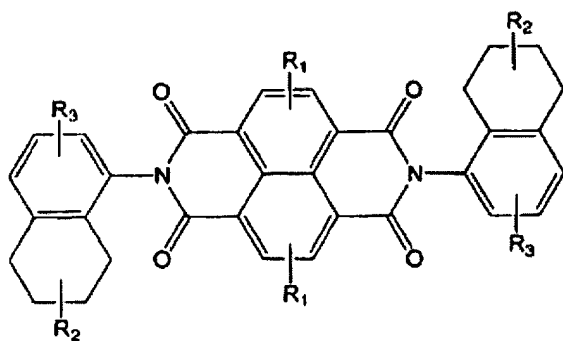
The naphthalenetetracarboxylic acid diimide derivative according to claim 1,  
 wherein the substituted alkyl, substituted alkoxy and substituted aralkyl are  
 independently substituted with any one selected from the group consisting of alkyl, aryl,  
 20 halogen and alkoxy.

## [Claim 3]

The naphthalenetetracarboxylic acid diimide derivative according to claim 1,  
wherein the substituted aryl is substituted with any one selected from the group  
consisting of alkyl, alkoxy, nitro and halogen.

**[Claim 4]**

5 An electrophotographic photoconductive material comprising a substrate and a  
photosensitive layer formed on the substrate, in which the photosensitive layer comprises a  
charge generating material and a charge transferring material, the charge transferring material  
is an electron transferring material and the electron transferring material comprises a  
naphthalenetetracarboxylic acid diimide derivative represented by the following formula:



10 in which, R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> are each independently any one selected from the group  
consisting of hydrogen; halogen; substituted or unsubstituted alkyl of C<sub>1</sub> to C<sub>20</sub>; substituted or  
unsubstituted alkoxy of C<sub>1</sub> to C<sub>20</sub>; substituted or unsubstituted aryl of C<sub>6</sub> to C<sub>30</sub>; and  
substituted or unsubstituted aralkyl of C<sub>7</sub> to C<sub>30</sub>.

15 **[Claim 5]**

The electrophotographic photoconductive material according to claim 4,  
wherein the substituted alkyl, substituted alkoxy and substituted aralkyl are  
independently substituted with any one selected from the group consisting of alkyl, aryl,  
halogen and alkoxy.

20 **[Claim 6]**



The electrophotographic photoconductive material according to claim 4,  
wherein the substituted aryl is substituted with any one selected from the group  
consisting of alkyl, alkoxy, nitro and halogen.

**[Claim 7]**

5           The electrophotographic photoconductive material according to claim 4, wherein the  
electrophotographic photoconductive material contains the electron transferring material of  
the above formula in a ratio of 20 wt% to 40wt% based on the total solid components.

**[Claim 8]**

10           The device as claimed in claim 7, wherein the first and the second guide rails  
comprise a receiving recess receiving the first and the second guide protrusions.

**[Claim 9]**

The electrophotographic photoconductive material according to claim 4,  
wherein the photosensitive layer has a single-layer structure comprising the charge  
generating material and the charge transferring material dispersed in an binder resin.

15           **[Claim 10]**

The electrophotographic photoconductive material according to claim 4,  
wherein the photosensitive layer has a layered structure comprising a charge  
generating layer comprising the charge generating material and a charge transferring layer  
comprising the charge transferring material.

20           **[Claim 11]**

The electrophotographic photoconductive material according to claim 4, wherein the  
electrophotographic photoconductive material further comprises an electron acceptor.